

Multilayer Film Assembly of Carbon Nanotubes

Alan M. Cassell*, M. Meyyappan, and Jie Han*

NASA Ames Research Center

Moffett Field, CA 94035

Key Words: Carbon nanotube, chemical vapor deposition, multilayer assembly

Abstract

An approach to assemble multilayers of carbon nanotubes on a substrate is presented. Chemical vapor deposition using a transition metal catalyst formulation is used to grow the nanotubes. Results show a bilayer assembly of nanotubes each with a different density of tubes.

Introduction

Carbon nanotubes (CNT) exhibit extraordinary mechanical properties and unique electronic properties which has led to an active research field focusing on nanoscale devices [1], sensors [2] and composites [3]. A key to the realization of these applications is the development of reliable, reproducible and economical growth techniques. The early CNT growth efforts centered primarily on laser ablation [4] and arc production [5] approaches. Though these

* ELORET Corporation

In our current work, catalyst formulations and reaction conditions have been developed to enable formation of multiple layers of nanotube films where the density of nanotubes grown from each layer could be controlled by choosing appropriate catalysts. A schematic of the processing sequence is shown in Fig. 1. First, 5 μL of precursor A (0.5 M SiCl_4 , and 6 mM P-123 tri-block copolymer in a 2:1 mixture of ethanol/methanol) was spun onto a 1-cm^2 N-doped silicon substrate ($\langle 100 \rangle$, $1\text{-}10\ \Omega\text{-cm}$) at 6000 rpm. Next, 5 μL of catalyst precursor B (0.5 M $\text{SiCl}_4/\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and 6 mM P-123 in a 2:1 mixture of ethanol/methanol) was spun on top of film A at 6000 rpm. The layered films were then annealed in a drying oven for 1 h at $100\ ^\circ\text{C}$, followed by further annealing at $700\ ^\circ\text{C}$ in air to remove the tri-block copolymer and render the catalyst layers active. The prepared substrate was then placed in the CVD reactor and exposed to argon flow at a rate of 500 sccm and $700\ ^\circ\text{C}$ for 30 minutes. Finally, nanotube growth was facilitated by introducing 500 sccm of ethylene at $750\ ^\circ\text{C}$ for 30 minutes.

The samples were examined in a scanning electron microscope (SEM) and the images are shown in Fig. 2. We see two distinct layers of nanotubes corresponding to the two films A and B discussed above. These are multiwall nanotubes in both layers. We note that layer B was pushed upward by nanotubes growing from layer A, leading to the observed nanotube bilayer. A close inspection of the image reveals that nanotubes grown from layer A (bottom layer) are lower density than nanotubes grown from layer B (top layer). We recall that layer A does not

multilayers for future spacecraft. For example, the outside high density layer can aim at self-sensing and adapting to the chemical, thermal and mechanical environment; the inner low density layer can be designed to allow high aspect ratio conducting tubes to be dispersed in dielectric hosts for effective electromagnetic shielding and also to promote sensing and healing to microcracks. The scaleup and incorporation of additives and host materials to the nanotube multilayers for enhanced performance are areas of our current research.

Nanotube multilayers also offer higher functionality for many other applications. They can be used as an effective armor with alternative low density layers; size-selective channels for molecular separation, drug delivery and microfluidics with different density layers; and templates for sensors, actuators, and biologically inspired structures with the incorporation of bioactive or other additives. They may also enable assembly of molecular electronics, optics and magnetics in a multilayer architecture of solid state devices. For instance, a nanotube multilayer with distinct functionality assigned to each layer may function as molecular electronics logic gates, as does a hybrid multilayer of organic molecules and solid state materials.

Acknowledgement

This work was supported by the NASA Ames Nanotechnology Program. The authors acknowledge the technical support of Sunita Verma.

8. Cassell A.M., Franklin N.R., Tombler T. W., Chan E.M., Han J., and Dai H., 'Directed growth of free-standing single-walled carbon nanotubes,' J. Am. Chem. Soc. **121**, 7975-7976 (1999).
9. Kind H., Bonard J. M., Forro L., Kern K., Hernadi K., Nilsson L., and Schlapbach L., 'Printing gel-like catalysts for the directed growth of multiwall carbon nanotubes,' Langmuir **16**, 6877-6883 (2000).
10. Cassell A.M., Verma S., Delzeit L., Meyyappan M., and Han J., 'Combinatorial optimization of heterogeneous catalysts used in the growth of carbon nanotubes,' Langmuir (2000).

Figure Captions

1. Schematic of the processing sequence
2. SEM image of the two layers of nanotubes. Figure on the right is a high magnification image.

